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CHAPTER–1
Introduction

1.1 GENERAL INTRODUCTION

Particulate matter (PM) generated during combustion and occurring in different size fractions, is a common component of ambient air (**Chuang et al., 2011**). Epidemiological studies have linked that exposure to ambient PM is associated with adverse health effects (**Dockery and Pope, III, 1996**), which is considered as a risk factor for cardiovascular morbidity (**Dominici et al., 2006**), and mortality (**Pope, III et al., 2004**). During burning, in addition to the PM emission, organic compounds are released as gases, and the combination or synergistic effects of the two different phases may increase the risk of pulmonary disease (**Polichetti et al., 2009**). Recently, combustion related activities have been the focus of inhalation toxicological and exposure assessments (**Mishra, 2003; Slezakova et al., 2009; Baumgartner et al., 2011; Chowdhury et al., 2012**); these include incense, candles and biomass burning. Biomass burning can be generally defined as open or quasi-open combustion of any non-fossilized vegetative or organic fuels (**Akagi et al., 2011**). It includes: 1) open fires in forest, savannas, agricultural waste burning, municipal waste management in outdoor environments; 2) biofuels burning (wood, cow dung, and crop residue for cooking and heating purpose); and 3) different types of customary practices in indoor/semi-indoor environments. These activities are considered to be potential major sources of toxic releases (**Andreae and Merlet, 2001; Akagi et al., 2011; Oahn et al., 2011; Yokelson et al., 2011; Zhang et al., 2013**). Nowadays, all combustion related indoor activities become a public health concern due to people spending the majority of their time indoors, which can contribute to poor indoor air quality (IAQ) (**USEPA, 2001; Taneja et al., 2008; WHO, 2010**). Nevertheless, burning incense and candle is a long-lasting tradition in many Asian countries including India as a common practice for religious and spiritual purposes and to give respect to ancestors for deity worship in Buddhism and Taoism. These rituals are commonly observed in altars at homes or in temples that are located in indoor environments (**Fine et al., 1999; Lin et al., 2007; Chuang et al., 2011**). Public concern over IAQ has increased strongly in recent years; hundreds of pollutants from different indoor and outdoor sources have been identified in indoor environments (**WHO, 1987**). Incense and candle

burning is one of the significant indoor sources of PM, trace gases such as carbon monoxide (CO), carbon dioxide (CO_2), oxides of nitrogen and sulfur (SO_x and NO_x), volatile organic compounds (VOCs), heavy metals, polycyclic aromatic hydrocarbons (PAHs), and speciated carbonaceous fractions (SCFs) (**Lee and Wang, 2004; Sun et al., 2006; Lin et al., 2007; Wang et al., 2007; Yang et al., 2007**). Previous study has shown that temple-indoors have a higher level of PM concentration than respective outdoors, which is a result of incense burning (**Lung et al., 2004**). **Lung et al.** (**1999**) has shown that 40% of 121 Taiwan households worship twice a day and are exposed to high levels of PM air pollution. Epidemiological investigations have demonstrated a relationship between burning incense and lung cancer (**Maclennan et al., 1977**), brain tumors (**Preston-Martin et al., 1982**), and childhood leukemia (**Lowengart et al., 1987**).

1.2 Review of the Work

Many laboratory and field based studies were conducted to determine the emission characters of PM and their chemical constituents emitted from incense, candles and biomass burning in various parts of the world. **Hays et al.** (**2005**) has characterized the physical and chemical composition of $PM_{2.5}$ (particulate matter with aerodynamic diameter ≤2.5 μm) emissions from the simulated agricultural fires involving rice and wheat residuals under aerobic conditions and CO/CO_2 ratio typical of field fires. The mean emission factors (EFs) of $PM_{2.5}$ from rice and wheat residual burning were found to be 13.0 $g.kg^{-1}$, and 4.7 $g.kg^{-1}$ respectively. In case of rice burning PM emissions, carbonaceous matter was the major component, whereas potassium (K) and chloride (Cl^-) were the major components of PM emissions resulting from wheat residue burning emissions. On characterizing the $PM_{2.5}$ solvent extracts, associated organic matter is accounted for about 18% of the particulates. In another study on characterization of gas emissions from an Amazonian forest clearing fire, CO_2, CO, and hydrocarbons have shown major dominance (**Soares Neto et al., 2009**). The average EFs obtained from the study were to be 1599 $g.kg^{-1}$, 111.3 $g.kg^{-1}$, 9.2 $g.kg^{-1}$, 5.6 $g.kg^{-1}$, and 4.8 $g.kg^{-1}$ for CO_2, CO, CH_4, Non methane hydrocarbon (NMHC), and $PM_{2.5}$ respectively.

Field and laboratory hood experiment were performed to investigate the emission characteristics of rice straw burning in Thailand. $PM_{2.5}/PM_{10}$, organic carbon (OC), elemental carbon (EC), metals, levoglucosan, methoxy phenol, and PAHs were found to be most abundant component of emissions (**Oahn et al, 2011**). **Cao et al.** (**2008**) reported the EF

estimates for PM, EC, OC, SO_2, NO_x, CO, CO_2 and ions from simulated cooking conditions using rice straw, wheat straw, corn stover, and cotton stalk in a combustion tower designed to replicate the brick stoves used in rural China that use crop residues as fuels. Results have shown that wheat straw had the highest EF for the total PM (8.75 $g.kg^{-1}$) among the four crop residues. Corn stover and wheat straw burning emissions have shown highest EF for EC (0.95 $g.kg^{-1}$), and OC (3.46 $g.kg^{-1}$), respectively. **Sahai et al.** (**2007**) performed an in situ experiment of wheat straw burning to develop specific EFs for India and to address the information gap on field burning of crop residue in the South Asian region. EFs were established and from which total emissions of CH_4, CO_2, CO, N_2O, NO, NO_2, and free carbon were estimated. Later, **Saud et al.** (**2011**) have presented the experimentally determined EFs of PM and trace gases (SO_2, NO, and NO_2) from biomass fuels used as residential fuels (dung cake, fuel-wood, and crop residue) in rural sector of Indo-Gangetic Plain in India. Results have established that the EFs of PM, SO_x, and NO_x have different pattern of emissions depending on fuel quality and location. Budget estimates of PM, SO_x, and NO_x from biomass fuel over IGP were also accounted, which were 41%, 22%, and 18%, respectively of total emission from India.

Emissions of PM from incense were measured for 23 different types of incense in a study carried out by **Jetter et al.** (**2002**), which was also focused on determination of emission rates (ERs) of CO, SO_2, and NO in a laboratory setting. $PM_{2.5}$ ERs were found to be in the ranges from 7 to 202 $mg.h^{-1}$. $PM_{2.5}$ EFs were found in the ranges from 5 to 56 $mg.g^{-1}$ of incense burned. The investigation of the particle size distribution, emission characteristics and profiling of PAHs from burning of nine types of incense was performed by **Yang et al.** (**2007**) using a custom-designed combustion chamber. The average range of EFs of S-PAHs (solid phase-PAHs) and G-PAHs (gas phase-PAHs) were found to be 4.52-6.93 $\mu g.g^{-1}$, and 10.6-29.8 $\mu g.g^{-1}$ incense, respectively. However, the particle-phase BaPeq [benzo(a)pyrene equivalent concentration] EF was found to be consistently more than 40-folds higher than that of the corresponding gas-phase BaPeq. These results clearly indicate that in terms of carcinogenic potency, S-PAHs are more carcinogen that G-PAHs. The correlation of metallic contents of incense to its burning rate and emission products was reported by **Lin et al.** (**2007**). Results have shown that, when the total metallic content (with Ca as the major component) was lower than 2% of the raw materials can effectively reduce the level of particulates (about 40%) during the combustion. **Chiang and Liao** (**2006**) studied the

assessment of human exposure to airborne PMs and PAHs from heavy incense burning in temples using a mechanistic-based exposure and risk models, appraised with reported empirical data. Results implicate that exposure to smoke emitted from heavy incense burning may promote lung cancer risk. The study provides a quantitative basis for objective risk prediction of heavy incense burning exposure in temples. **Wang et al.** (**2007**) conducted field investigations of target air pollutants from two temples in Hong Kong including $PM_{2.5}$ and PM_{10}, VOCs, carbonyl compounds, CO, NO_x, CH_4, NMHC, OC, EC, and inorganic ions (Cl^- ,NO_3^-,SO_4^{2-},Na^+,NH_4^+, and K^+). In general, the average $PM_{2.5}/PM_{10}$ ratio was approximately 82%. $PM_{2.5}$ constituted the majority of suspended particulates at both temples. It was noted that formaldehyde was the most abundant carbonyl compounds, followed by acetaldehyde. During peak burning period, average benzene concentration exceeded almost eight times more than the Hong Kong IAQ standard.

See and Balasubramanian (**2011**) reported EF of $PM_{2.5}$ and its chemical constituents emitted from six different brands of incense sticks in a controlled experiments. Results have shown that $PM_{2.5}$ EFs range from 0.4 (smokeless incense stick) to 44.5 $mg.g^{-1}$. It was also found that the amount of metals emitted is highly dependent on the quantity of metals present in the incense sticks. **Sun et al.** (**2006**) conducted a series of candle-burning experiments in a well-controlled test chamber, and monitored the characteristic of size distribution, number and mass concentration using a scanning mobility particle sizer (TSI, Model SMPS-3034). The number emission values of candle smoke particles of the steady burn, unsteady burn and smoldering modes were found to be $4.05\times10^{12}\pm0.73\times10^{12}$ $units.g^{-1}$, $1.49\times10^{12}\pm0.32\times10^{12}$ $units.g^{-1}$, and $1.55\times10^{11}\pm2.29\times10^{10}$ $units.g^{-1}$, respectively, and the corresponding PM_{10} EFs were found to be 0.055 ± 0.004 $mg.g^{-1}$, 1.08 ± 0.32 $mg.g^{-}1$, and 0.24 ± 0.07 $mg.g^{-1}$, respectively. **Pagels et al.** (**2009**) also investigated the physical and chemical properties of particle emissions from candle burning in indoor air. The large differences in composition, particle size and effective density between the different modes in the particle size distribution of candle emissions illustrate that distinctly different particle types were emitted during the different modes of candle burning. This has important implications for potential adverse health effects.

1.3 DEVELOPMENT OF EMISSION FACTORS AND EMISSION ESTIMATION

EMISSION FACTOR

A consideration commonly used to describe emissions from different types of burning is the EF, generally expressed in units of $g.kg^{-1}$. It is calculated by dividing the mass of emission by the mass of the dry material consumed by fire (**Andreae and Merlet, 2001**). Mathematically, EFs are generally calculated by stoichiometric considerations using the Carbon Mass Balance Method with explicit equations (**Hurst et al., 1994; Delmas et al., 1995; Yokelson et al., 1999; Dhammapala et al., 2006; Dhammapala et al., 2007a, 2007b**).

$$EF_i\left(g.kg^{-1}\right) = \frac{\triangle C_i \times 1000\left(g.kg^{-1}\right) \times C_{fraction}}{\left(\triangle CO_2 - C + \triangle CO - C + \triangle THC - C + \triangle PM - C\right)}$$

Where, EF_i = emission factor of pollutant i in $g.kg^{-1}$,
$\triangle C_i$ = background-corrected concentration of pollutant i in $mg.m^{-3}$,
$\triangle CO_2$-C = mass fraction of C in CO_2 in $mg.m^{-3}$,
$\triangle$CO-C = mass fraction of C in CO in $mg.m^{-3}$,
$\triangle$THC-C = mass fraction of C in total hydrocarbon in $mg.m^{-3}$,
$\triangle$PM-C = mass fraction of C in particulate matter in $mg.m^{-3}$,
$C_{fraction}$ = carbon content in dry material.

The THC and PM carbon are typically <<1% of the $CO+CO_2$ and can be neglected (**Ward, 1991; Ward et al., 1992**). EFs have been used for various applications, providing an easy estimation of ERs and concentration of emitted pollutants and are of great significance in developing national environmental policies and protection strategies**.**

EMISSION ESTIMATION

The method used for the emission calculation for the species i is based on the following equation given by **Seiler and Crutzen** (**1980**) **and Kanokkanjana et al.** (**2011**).

$$E_i = M \times EF_i$$

Where E_i = emission flux of pollutant i ($Gg.yr^{-1}$), M= amount of dry material burned (kg), EF_i= emission factor given for the pollutant i in grams per kilogram of dry matter.

1.4 RESEARCH HYPOTHESIS

The research conducted here stems from the following hypotheses:

Real-world burns are hypothesized to provide reasonable representations compared to those of chamber burns. Most laboratory scale trials described in the literature seem to consist of the combustion and sampling of emissions. However, it is hypothesized that real-world burns would have combustion characteristics different from those of chamber burns, owing to differing degrees of contact between the materials and available oxygen. It is hypothesized that EF evaluation of those carbonaceous matters which represent major components of particle- and gas-phase emissions from real-world biomass burning in different major ritual/cultural practices will provide realistic emission estimates of various hazardous carbonaceous pollutants. These estimates will be useful in IAQ assessment of various ritual/cultural activity centers.

1.5 OBJECTIVES OF THE PRESENT WORK

The goal of the study is to estimate the emissions of selected carbonaceous pollutants emitted from different types of burning activity in ritual and cultural centers.

The specific objectives of the study are:

1. To develop real-world EFs and to estimate annual emission budget of selected particulate- and gaseous organic air pollutants (temperature resolved sub-fractions of OC & EC, P-PAHs, and VOCs) emitted from ritual/cultural burning practices.
2. To measure IAQ of major ritual/cultural-indoors/semi-indoors for selected particulate- and gaseous organic air pollutants.
3. To investigate seasonal & temporal trend and indoor/outdoor (I/O) relationships associated with selected indoor organic air pollutants in ritual/cultural centers.
4. To investigate fireworks' emission markers and their enrichment in ambient $PM_{2.5}$ during fireworks festival Diwali.

1.6 Scope of the Present Work

Most of the studies being conducted in the developing countries are mainly focused on emissions from incense and candles burning activities. EFs of air pollutants from real-world natural and synthetic biomass burning during ritual and cultural practices were not well characterized. In addition, there is a lack of information on comparison of types and quantity of air pollutants emitted from different kinds of material burning and IAQ in ritual and cultural places. Thus, the present investigations will provide a systematic information regarding emissions of carbonaceous pollutants from ritual and cultural sites and also will serve as a tool for air quality management and emission inventory, concerning smoke emissions from ritual and cultural based combustion in India.

IAQ assessment data, so obtained, in this study will provide useful information regarding current status of $PM_{2.5}$/PM_{10}, OC & EC fractions, 14 VOCs, and 70 P-PAHs. It will help to identify possible mitigation practices for PM reduction in major ritual/cultural centers located in the South and Southeast Asia. The emission estimates of indoor PM concentration and associated carbonaceous fractions, and P-PAHs along with VOCs, evaluated in the study is an useful database for further risk assessment or health-related studies as well as regulatory agencies to set up guidelines and setting up standards for indoor ritual and cultural places.

CHAPTER–2
Methodology

A. AREA OF THE WORK

Raipur City, District Raipur, Chhattisgarh

B. STUDY DESIGN

The study was mainly focused on development of EFs for RPM (particulate matter with aerodynamic diameter ≤5 µm), OC, EC, P-PAHs along with VOCs in emissions resulting from ritual and cultural based burning activities. Following five different types of ritual and cultural sites have been chosen:

1) Hindu Temples (HT);

2) Muslim Holy Shrines (MHS);

3) Buddhist Temples (BT);

4) Marriage Places (MP); and

5) Hindu Cremation Centers (CC)

Study was designed in such fashion that real-world EFs of major carbonaceous pollutants (OC, EC, P-PAHs, and VOCs) during biomass burning activities involved with major ritual/cultural practices had to be determine using standard reported procedures and the EFs database had to be use in annual budget estimation of selected carbonaceous pollutants. Consequently, similar examples of selected ritual/cultural-indoors/semi-indoors were planned to monitor their respective IAQ for similar organic pollutants with respect to investigate seasonal/temporal trend and I/O relationship. Pure residential-indoors (RESID-indoors) and one common ambient-outdoor (AMB-OUT) were also selected to measure organic pollutants to compare them with those measured in ritual/cultural-indoors/semi-indoors. A stratified random sampling design using longitudinal measurement plan has been adopted for monitoring of indoor air $PM_{2.5}$/PM_{10}, P-PAHs, carbonaceous fractions (OC & EC), and VOCs in selected community activities (**Gilbert, 1987; USEPA, 2003**).

C. SAMPLING DESIGN

(I) Sampling Plan for Development of EFs

Sampling plans for development of EFs for RPM, OC, EC, P-PAHs, and VOCs were designed according to standard protocol presented earlier (**Delmas et al., 1995; Lee and Wang, 2004**). Three separate examples of each of these five ritual and cultural venues were sampled when materials were burned during events. Concentrations of RPM, CO_2, CO, and VOCs were sampled from the smoke plumes.

RPM SAMPLE COLLECTION

Three handy samplers (Envirotech Model APM 821) equipped with a 5 µm 50% cut-point cyclone inlets were used in each sampling event to determine field uncertainties associated with mass concentrations. RPM was sampled onto prebaked quartz fiber filters (QFFs) (47 mm diameter, Whatman QM/A) attached to handy samplers at an average flow rate of 2.0 lpm with reported protocol of sampling (**Dhammapala et al., 2006; Dhammapala et al., 2007a, 2007b; Oahn et al., 2011**). Individual RPM samples for each burn were typically collected over the entire burn duration. A total of 71 RPM samples (MP=18; MHS=12; BT=8; HT=26; and CC=7) were collected at ritual/cultural sampling sites. Similar sampling program of RPM was also carried out before the start of burning activities in all events of sampling for background correction. Field blank levels were also determined in the same way before the start of burning activities, averaged and subtracted from the sample measurements while propagating the standard deviation of the average blanks to compensate for adsorbed organic vapours on the QFFs (**Watson et al., 2009; Chow et al., 2010**).

VOCS SAMPLE COLLECTION

VOCs emitted during burning of material in selected ritual and cultural places were passively collected using Radiello® VOC samplers (Supelco Analytical). VOC passive samplers were installed above the smoke plume at different heights according to the recommended sampling protocol (**Lee and Wang, 2004; Radiello, 2010; Mason et al., 2011**). VOC samples for each burn were normally collected over the entire burn period. A total of 71 VOCs samples (MP=18; MHS=12; BT=8; HT=26; and CC=7) were collected at ritual/cultural sampling sites. For the background corrections, passive samplers were positioned in the same site/same height and for same duration before the start of burning activities. In each sampling event, three passive samplers were installed to determine field uncertainties associated with VOCs concentration.

GASEOUS POLLUTANTS

Simultaneously, in plume CO and CO_2 concentrations were measured with a portable NDIR monitors, (Langan Products Inc., San Francisco, CA, Model T15v and AZ Instrument Corporation, Taiwan, Model AZ 7755, respectively). Measurements have also been carried out before the start of burning activities for background corrections. All procedures such as sample collection, handling of filters, transportation, and preservation have been conducted using reported protocol elsewhere (**Lee and Wang, 2004; Hays et al., 2005**).

(II)Sampling Plan for IAQ Assessment

$PM_{2.5}$/PM_{10} SAMPLE COLLECTION

Same ritual/cultural-indoors/semi-indoors were again monitored for IAQ in relation to assess the indoor levels of those selected organic pollutants which were taken into account during EFs development. Apart from, pure-RESID-indoors and one common AMB-OUT site were also chosen to compare those found in ritual/cultural-indoors/semi-indoors. Only difference between these two sampling objectives (EF development and IAQ assessment) is the location of samplers and duration of sampling. In case of IAQ assessment, samplers were not operated over the smoke plume released from biomass burning, but positioned in a random place within the indoor/semi-indoor environment. A set of three units of fine particulate samplers (Envirotech, Model APM 550) at air flow rate of 16.7 lpm were used for monitoring of each of $PM_{2.5}$ and PM_{10} in all selected indoors/semi-indoors and ambient-outdoor environments. Set of three units of $PM_{2.5}$/PM_{10} samplers were used in each sampling event to determine field uncertainties associated with mass concentrations. Accordingly, each sample is a set of three sampled QFFs. A 24 h measurement of $PM_{2.5}$/PM_{10} has been carried out throughout the sampling year (March 2012-February 2013) using standard protocol of sampling (**Envirotech, 2000; USEPA, 2003; Kulshrestha et al., 2009**). Mass concentration of $PM_{2.5}$ and PM_{10} were collected onto prebaked QFFs. Laboratory and field blanks were also determined using recommended procedures (**Watson et al., 2009; Chow et al., 2010**). A total of 432 samples (having three sampled QFFs) have been collected, comprising of 72 samples, consisting 36 $PM_{2.5}$ and 36 PM_{10} samples from each cultural/ritual-indoors/semi-indoors along with RESID-indoors during all three seasons (fall, winter and summer) to investigate seasonal variation. Total number of AMB-OUT samples was 24 (having three sampled QFFs) comprising 12 $PM_{2.5}$ and 12 PM_{10} samples, respectively; once in each month throughout the one calendar year (March 2012-February 2013) to determine I/O relationship. Standard

procedures have been used for handling and operation of samplers (**Chow and Watson, 1998**).

VOCs Sample Collection

A Radiello® diffusive sampler has been used for collection of VOCs at designated indoor/semi-indoors sampling sites. A 48 h period (except RESID-indoors for 4 days sampling period) measurements of VOCs has been carried out throughout the sampling year (March 2012-February 2013) using recommended procedure of sampling (**Bruno et al., 2008; Radiello, 2010; Mason et al., 2011**). A total of 216 VOC samples (having three sampled cartridges) were collected, consisting 36 VOC samples from each cultural/ritual-indoors/semi-indoors along with RESID-indoors during all three seasons (fall, winter and summer) to investigate seasonal variability. Total number of AMB-OUT VOCs samples was 12 (having three sampled cartridges), once in each month throughout one calendar year (March 2012- February 2013) to determine I/O relationship.

D. CHEMICAL ANALYSIS

Analysis of Carbonaceous Fractions & Quality Control

The mass of RPM, $PM_{2.5}$ and PM_{10} have been determined using standard gravimetric method. Sampled QFFs were subjected to chemical analysis of temperature resolved carbonaceous fractions using thermal optical reflectance (TOR) procedure using DRI model 2001 carbon analyzer by following IMPROVE_A TOR protocol (**Chow et al., 1993; Chow et al., 2004; Chow et al., 2007**). This analysis produced four OC fractions (OC1, OC2, OC3, and OC4); OP (a pyrolyzed carbon); and three EC fractions (EC1, EC2, and EC3). IMPROVE_TOR OC is operationally defined as OC1+OC2+OC3+OC4+OP and EC is defined as (EC1+ EC2+EC3)−OP (**Chow et al., 2007**). In addition to IMPROVE_A protocol, NIOSH 5040 protocol was also employed using semi-continuous thermal/optical carbon analyzer (Sunset Laboratory Inc. Model 4L) to quantify OC and EC fractions associated with $PM_{2.5}$ and PM_{10} measured during IAQ monitoring (**Birch and Cary, 1996**). The quality assurance and quality control for carbonaceous fractions analysis have been conducted by using standard protocol (**Chow et al., 1993; Birch and Cary, 1996**).

Analysis of P-PAHs & Quality Control

Another sampled QFFs of RPM and $PM_{2.5}$/PM_{10} were used for determination of P-PAHs using standard procedures **(Zielinska et al., 2004; DRI, 2010; He et al., 2010**). Extracted samples were then subjected to chemical analysis of selected seventy P-PAHs using Gas Chromatography/Mass Spectrophotometer

using standard procedures and recommended conditions of operation (**Zielinska et al., 2004; DRI, 2010; He et al., 2010**). Description of seventy P-PAHs was, so measured, given in **Table 2.1**. Measured P-PAHs were grouped in total P-PAHs (TP-PAHs), heavy P-PAHs (HP-PAHs), and light P-PAHs (LP-PAHs). The concentrations of TP-PAHs are defined as the sum of the concentrations of the measured 70 PAHs species for particle phase. To understand the distribution of PAHs homologues for each collected sample, P-PAHs were classified into two categories according to their molecular weights: light P-PAHs having low molecular weight PAHs (LP-PAHs-containing 2 to 3-ring PAHs), and heavy P-PAHs having high molecular weight PAHs (HP-PAHs- containing 4-7-ring PAHs) (**Mu et al., 2013**).The quality assurance and quality control for P-PAHs analysis have been conducted by using standard protocol (**Zielinska et al., 2004; DRI, 2010; He et al., 2010**).

Table.2.1: List of Measured P-PAHs

Particulate Bound Polycyclic Aromatic Hydrocarbons		
Light PAHs (LP-PAHs= 25)	**2- Ring**	2-phenylnaphthalene
	3- Ring	A-Methylfluorene, B-Methylfluorene, 1-Methylfluorene, 9-fluorenone, Dibenzothiophene, Phenanthrene, Anthracene, 3-methylphenanthrene, 2-methylphenanthrene, 2-methylanthracene, 4,5-methylenephenanthrene, 9-methylphenanthrene, 1-methylphenanthrene, 9-methylanthracene, A-dimethylphenanthrene, B-dimethylphenanthrene, 1,7-dimethylphenanthrene, 3,6-dimethylphenanthrene, D-dimethylphenanthrene, E-dimethylphenanthrene, C-dimethylphenanthrene, Retene
Heavy PAHs (HP-PAHs= 45)	**4- Ring**	Fluoranthene, Pyrene, benzo(a)fluorine, benzo(b)fluorine, 5-methylchrysene, 1+3-methylfluoranthene, 4-methylpyrene, 1-methylpyrene, Benzonaphthothiophene, benzo(c)Phenanthrene, 9-phenylanthracene, 6-methylchrysene, Benz(a)anthracene, Chrysene-Triphenylene, Benzanthrone, Benz(a)anthracene-7,12-dione, 3-methylchrysene, 7-methylbenz(a)anthracene, 7,12-dimethylbenz(a)anthracene
	5- Ring	Benzo(ghi)fluoranthene, Cyclopenta(c,d)pyrene, Benzo(b)fluoranthene, Benzo(j)fluoranthene, Benzo(k)fluoranthene, Benzo(a)fluoranthene, BeP, BaP, Perylene, 7-methylbenzo(a)pyrene, 9,10-dihydrobenzo(a)pyrene-7(8H)-one, dibenz(a,h)acridine, dibenz(a,j)acridine, Dibenzo(ac)anthracene, Dibenzo(ah)anthracene, Dibenzo(a,j)anthracene, Benzo(b)chrysene, Picene
	6- Ring	Indeno[123-cd]fluoranthene, Indeno[123-cd]Pyrene, Benzo(ghi)perylene, Anthanthrene, Dibenzo(a,l)Pyrene, Dibenzo(a,e)Pyrene, Dibenzo(a,i)Pyrene, Dibenzo(a,h)Pyrene, Dibenzo(b,k)fluoranthene
	7- Ring	Coronene

ANALYSIS OF VOCS & QUALITY CONTROL

All Radiello samples were analyzed by the thermal desorption cryogenic preconcentration method, followed by high-resolution gas chromatographic separation and mass spectrometric detection of individual compounds using standard procedures and recommended conditions of operation (**DRI, 2010; Mason et al., 2011**). Fourteen VOCs namely — n-hexane, cyclohexane, n-heptane, n-octane, n-nonane, n-decane, n-undecane, styrene, o-xylene, m,p-xylene, 1,2,4-trimethyl benzene, ethyl benzene, benzene, and toluene were measured in ritual and cultural places which were hazardous to humans according to USEPA Air Toxic Programme (**USEPA, 1994**). Total VOCs (TVOCs) were calculated by summation of the 14 individual VOCs concentrations for each venue. The quality assurance and quality control for VOCs analysis have been conducted by using standard protocol (**DRI, 2010; Mason et al., 2011**).

E. DATA ANALYSIS AND DOCUMENTATION

Data, so obtained, in compliance to the objectives of development of EFs of selected organic pollutants (RPM, OC, EC, P-PAHs, VOCs, CO_2, and CO), were documented with annual budget of emission estimates of respective species and comparison with earlier reported values (**Dhammapala et al., 2006; Kanokkanjana et al., 2011**). IAQ monitoring data have been documented using annual mean and their respective standard deviation to compare with IAQ standards of WHO and earlier reported values and those obtained for RESID-indoors and AMB-OUT using boxplot statistical graphs. Seasonal and temporal variation, I/O relationships using regression analysis and $PM_{2.5}/PM_{10}$ ratios for associated OC, EC, and P-PAHs were also evaluated and documented. All the statistical calculations and plots were carried out using MS Excel 2010 and Origin 8.5 software.

CHAPTER–3

Volatile Organic Compounds in Indoor and Semi-Indoor Ritual Places: Emission Factors, Annual Trend and Indoor/Outdoor Relationship

VOCs are an important group of organic compounds which plays an important role in several photochemical processes, the global carbon cycle, influence the budget of tropospheric O_3, and small direct impact on radiative forcing (**Fehsenfeld et al., 1992**). Recent estimates of the global emission of VOCs into the atmosphere range from about 1200 to 1600 $Tg.yr^{-1}$ (**Reimann and Lewis, 2007**). Asian emissions of VOCs are found to be 68.5 $Tg.yr^{-1}$, in which 39.5% share comes from China (27.1 $Tg.yr^{-1}$), and Indian share was 23.3% (15.95 $Tg.yr^{-1}$) (**Kurokawa et al., 2013**). Anthropogenic activity sectors wise contribution assessment has shown that 43% comes from domestic biofuels consumption, whereas road transport sector contributes 35% to the total VOCs emissions in India (**Kurokawa et al., 2013**). Recently, it was estimate that Indian annual emission budget of VOCs was 100-470 $Gg.yr^{-1}$ from open biomass burning activities (**Pandey and Sahu, 2014**). It has also been reported that incense/candles burning are significant sources of VOCs, which have direct impact on environments (**Lee and Wang, 2004**). Emissions of VOCs from biomass burning activities involved in major ritual/cultural places in India are not yet documented. Thus, the present study estimates real-world VOCs emission during biomass burning activities involved with major ritual/cultural practices in contrast to other studies that examined emissions in laboratory scale from specific types of incense or biomass material. Description of sampling sites, sampling protocol and methods adopted for chemical analysis has been given earlier in Chapter 2. It was observed that CC (229.17±39.18 $g.kg^{-1}$), and HT (0.09±0.01 $g.kg^{-1}$) have shown highest and lowest EFs of TVOCs. Benzene (0.24-33.22 $g.kg^{-1}$) has shown the highest EF among 14 VOCs in all places. MP, MHS and BT have shown different pattern of VOC EFs compared to laboratory-based controlled chamber studies. Based on the activity levels in ritual/cultural places in India (Base Year 2010), the annual TVOCs emission determined as 954.25 $Gg.yr^{-1}$ in which maximum VOCs emissions comes from CC (953.34±162.99$Gg.yr^{-1}$).

The result of this investigation has been facilitated to monitored IAQ in these ritual and cultural sites. IAQ monitoring protocols and methods adopted for chemical analysis have been described earlier in Chapter 2. The annual mean concentrations for TVOCs were found to be 216.61±75.15, 656.34±220.82, 681.75± 219.83, 129.51± 45.24, 60.95±19.48, 82.67± 40.96, and 34.15±13.52 $\mu g.m^{-3}$ for MP, MHS, BT, HT, CC, RESID, and AMB-OUT, respectively. Toluene, and m,p-xylene has been observed to be most abundant in all ritual/cultural-indoors compared to other VOCs whereas toluene and benzene have shown most abundant species in CC-semi-indoors. Most of the individual VOCs were found to be higher than the prescribed standards and earlier reported indoor VOCs levels. The order of occurrence of all individual and TVOCs in all ritual/cultural-indoors/semi-indoors were found as: winter>fall>summer; inversely related to climate temperature: summer>fall>winter. Regression analyses have shown that indoor contribution for most of VOC was ~51-85% in all ritual/cultural-indoors/semi-indoors, significantly higher than that for RESID-indoors (<45%). Higher I/O ratios and weak correlation, suggested that indoor generated sources have shown dominance over outdoor sources and indoor activities itself influenced VOCs concentrations.

CHAPTER–4

Speciated Carbonaceous Fractions in Indoor and Semi-Indoor Ritual Places: Emission Factors, Temporal Variation and Indoor/Outdoor Relationship

Particulate black carbon (BC, sometimes called EC), and OC affect climate, visibility, and human health. Recent estimates of global BC and OC emissions ranges from 8 to 24 $Tg.yr^{-1}$, and 33 to 62 $Tg.yr^{-1}$, respectively (**Chow et al., 2010**), whereas Asian BC and OC emissions accounted for 3.03 $Tg.yr^{-1}$, and 7.72 $Tg.yr^{-1}$ (**Kurokawa et al., 2013**). China (1.59, 3.08 $Tg.yr^{-1}$), and India (0.71, 2.29 $Tg.yr^{-1}$) were found to be largest emitters of BC and OC among the Asian countries. Biomass fuels were found to be major contributor of OC (797.56 $Gg.yr^{-1}$), and EC (177.40 $Gg.yr^{-1}$) emissions in India (**Saud et al., 2012**). It has also established that incense/candles burning are important sources of carbonaceous aerosols (**Lee and Wang, 2004; Pagels et al., 2009**). Carbonaceous aerosol emissions from biomass burning activities involved in major ritual/cultural places in India are not recognized. Multi-source contribution to the atmospheric carbonaceous aerosols reported earlier has attributed to the need of determination of EFs of temperature resolved speciated sub-fractions of carbonaceous matter (OC1, OC2, OC3, OC4, OP, EC1, EC2, and EC3) associated with RPM emitted from specific domestic biomass burning activities occur in major ritual and cultural places in India.

This chapter mainly focused on development of OC/EC EFs released from burning activities involved in major ritual/cultural places in India, which have significant implications on indoor- and outdoor air quality along with atmospheric chemistry involved. The major species CO_2, CO, and carbonaceous fraction associated with RPM have been identified by using specific standard protocols of sampling and analysis described earlier in Chapter 2. Range of average EFs of CO_2, CO, RPM, OC, and EC, were measured to be 1378.20-1783.25, 105.23-315.16, 20.13-163.02, 3.47-27.04 and 0.18-7.96 $g.kg^{-1}$ respectively. It was observed that OC1, major marker of biomass burning has shown higher EF in MHS, while OC2 has shown higher EF in BT. In contrast EFs of OC3 and OC4 fraction have shown higher in CC.OP; an indicator of water soluble organics has shown uniform EFs values in all sites. EC fractions, reported to be the fractional masses of soot (EC2 + EC3), and char (EC1 – OP) emissions have shown different EF values in HT compared to other sites. Char has been observed to be dominant in emissions resulting from burning practices involved with MP, MHS, BT and CC, whereas soot is predominant in case of HT. Total Indian carbonaceous

fractions emissions from these ritual/cultural sites were 166.18± 13.64 $Gg.yr^{-1}$, of which 116.35±10.76 $Gg.yr^{-1}$ was in the form of OC and 49.83±2.87 $Gg.yr^{-1}$was in the form of EC.

Standard protocol of IAQ measurement and chemical analysis has been adopted for SCFs analysis described earlier in Chapter 2. The annual mean concentration for $PM_{2.5}$ was found to be 1154.52±415.14, 1422.68±456.32, 671.48±234.70, 436.14±138.75, 189.18±62.53, 219.27±89.50, and 178.16±79.17$\mu g.m^{-3}$ and those for PM_{10} was 1491.29±516.20, 1816.07±495.93, 924.86±303.20, 591.09±205.12, 276.38±84.11, 308.44±117.97, and 255.55±113.77$\mu g.m^{-3}$ for MP, MHS, BT, HT, CC, RESID and AMB-OUT, respectively. It was observed that annual mean concentration of $PM_{2.5}/PM_{10}$ at selected ritual/cultural-indoors many times higher than RESID-indoor, AMB-OUT as well as WHO standard. Whereas CC-semi-indoors has shown comparable annual concentration of $PM_{2.5}/PM_{10}$ with RESID-indoors and AMB-OUT but many times higher than WHO standards. Relative strength of occurrence of SCFs have shown different pattern in ritual/cultural-indoors/semi-indoors compared to AMB-OUT. Similarly, carbonaceous fractions have shown many times higher at all ritual/cultural-indoors compared RESID-indoors and AMB-OUT. As concern of temporal variation, higher degree of seasonal variations in SCFs associated with both PM fractions have been observed compared to those found in RESID-indoors and AMB-OUT. OC-EC correlations at selected ritual/cultural-indoors/semi-indoors sites are strong (R^2>0.87-0.99). It implies that a majority of the indoor OC comes from emission source similar to those responsible of EC. Regression analyses have shown that indoor contribution for OC & EC associated with $PM_{2.5}/PM_{10}$ fractions have 55-78% in all ritual/cultural-indoors/semi-indoors, significantly higher than that for RESID-indoors (<40%). Higher I/O ratios and weak correlation, and strong OC-EC correlation suggested that indoor activities itself influenced SCFs concentrations.

CHAPTER–5

Particle Bound Polycyclic Aromatic Hydrocarbons in Indoor and Semi-Indoor Ritual Places: Emission Factors, Seasonality and Indoor/Outdoor Contribution

PAHs are of special interest due to their toxicity, carcinogenicity, and ubiquitous distribution in the environment. PAHs are emitted during incomplete combustion from domestic, industrial power plants, traffic, open biomass burning, and pyrolysis of fossil fuels, biomass, and coke production (**Xu et al., 2006; Ravindar et al., 2008; Zhang and Tao, 2009**). It was estimated that the total global atmospheric emission of PAHs was 530 $Gg.yr^{-1}$ with biofuel (56.7%), wildfire (17.0%), and consumer product usage (6.9%) as the major sources, and China (114 $Gg.yr^{-1}$), India (90 $Gg.yr^{-1}$), and United States (32 $Gg.yr^{-1}$) were the top three countries with the highest PAH emissions. Biofuel burning (66.4-92.5%) was the dominant PAH source in India and China (**Zhang and Tao, 2009**). It has also been reported that smoke released during burning of incense/candles are significant sources of PAHs, which have direct impact on environments as well as human health. A number of investigations on the ERs and EFs associated with the particulates and PAHs produced by burning incense/candles have been reported in developing countries (**Yang et al., 2007; Orecchio, 2011; Yang et al., 2012**), but there is no available PAHs EFs and emission inventory for biomass burning activities involved with ritual/cultural activities in India. Therefore, the main objective of this chapter was to develop EFs and annual emission budget estimation for P-PAHs for various types of burning activities in major ritual and cultural places in India. Standard protocol of sampling and chemical analysis have been adopted which is given earlier in Chapter 2. The EFs for TP-PAHs from MP, MHS, BT, HT and CC were estimated as 77.04±5.39, 99.09±6.02, 90.35±6.76, 22.78±1.57, and 77.15±6.16 $mg.kg^{-1}$, respectively. Higher levels of TP-PAHs were found from MHS as compared to other sites. Fluoranthene, Pyrene, 9-fluorenone, Phenanthrene, 2-methylphenanthrene, benzo[a]anthracene, Benzanthrone, Benzo(ghi)fluoranthene constituted the major fraction of P-PAH emissions from all sites. Country level emission budget of TP-PAHs from ritual and cultural burning practices has also been evaluated to 0.47 $Gg.yr^{-1}$; contributed ~23% of the 2.07 $Gg.yr^{-1}$ — Indian emission budget of PAHs from unknown sources reported earlier.

Standard protocol of IAQ measurements and chemical analysis have been adopted for monitoring of P-PAHs and described earlier in Chapter 2. Results have shown that annual mean concentration of TP-PAHs associated with $PM_{2.5}/PM_{10}$ at selected ritual/cultural-indoors was many times higher than RESID-indoor as well as AMB-OUT. Individual P-PAHs has shown different pattern of occurrence in ritual/cultural-indoors/semi-indoors. Significant seasonal variation has also been observed for LP-PAHs and HP-PAHs in both particulate fractions. Regression analyses have shown that indoor contribution for most of P-PAHs associated with $PM_{2.5}/PM_{10}$ fraction have ~55-80% in all ritual/cultural-indoors/semi-indoors, significantly higher than that for RESID-indoors (<40%). Higher I/O ratios and weak correlation, suggested that indoor generated sources have shown dominance over outdoor sources and indoor activities itself influenced P-PAHs concentrations.

CHAPTER–6

Ambient $PM_{2.5}$ during Festivals Involved Fireworks: Study of Mass Re-Construction and Source Signatures

This chapter was designed with special emphasis on ambient air quality during Diwali episode. Many studies have shown fireworks, emits huge amounts of smoke which, contain massive quantities of health affecting and dangerous air pollutants within a short period of time, are exclusive sources of atmospheric pollution (**Murty, 2000; Ravindra et al., 2003; Wang et al., 2007; Sarkar et al., 2010; Tsai et al., 2012**). This study has been planned to assess the impact of emissions resulting from cracker burning during Diwali Festival on mass and chemical characterization of ambient $PM_{2.5}$; it includes elemental, ionic, and carbonaceous components. A three-week long ambient air $PM_{2.5}$ monitoring program during November 2012 in Bhilai, District Durg, Chhattisgarh, India has been carried out in two pure residential sites with standard protocol of sampling (**Sarkar et al., 2010; Tsai et al., 2012**). 8 h sampling was started at 16:00 h in each of sampling day and continued till12:00 h mid night. Sampled QFFs of each sampling event were subjected to chemical analysis by using standard protocol (**Chriswell et al., 1986; Harrison and Perry, 1986; Chow et al., 1993; Chow and Watson, 1999; Allen et al., 2001; Chow et al., 2004; Chow et al., 2007; Thermo Fisher, 2008**). $PM_{2.5}$ were chemically characterized with respect to twenty one metallic elements (Al, As, Ca, Cd, Co, Cr, Cu, Fe, Hg, K, Mg, Mn, Mo, Na, Ni, Pb, S, Sb, Se, V, and Zn), nine water soluble ionic species (F^-, Cl^-, NO_3^-, SO_4^{2+}, Na^+, NH_4^+, K^+, Ca^{2+}, and Mg^{2+}), and eight carbon fractions of organic (OC1, OC2, OC3, OC4 & OP), and elemental carbon (EC1, EC2 & EC3).

The maximum concentration of atmospheric $PM_{2.5}$ was found on Diwali day (1501.20µg.m^{-3}) followed by 2nd day of Diwali (831.98 µg.m^{-3}) and day after 2nd day of Diwali (397.12µg.m^{-3}) ; 2-8 folds higher compared to normal days and 7-25 times higher than NAAQS standards. The concentrations of metallic elements on Diwali days were 2.1-37.6 times higher than those from pre-Diwali days. The most abundant ionic species of $PM_{2.5}$ were Ca^{2+}, Cl^-, K^+, SO_4^{2-}, and Mg^{2+}during Diwali firework days. The molar ratio of Cl^- to Na^+ was rose significantly up to 3.84 during firework periods compared to on normal day (1.85). Similarly, On Diwali day the anion/cation ratio was found close to factor of two 2 (1.8); higher than those of normal days (1.1), described the acidic character of $PM_{2.5}$ aerosols on Diwali days.

It was observed that EC levels rose up to 6.4-folds from normal days to Diwali days and then dropped down moderately in post-Diwali days. Similarly, OC is increased on Diwali days by a factor of 5.9 compared to those measured in normal days. As far as SCFs of OC and EC is concern, nearly uniform association with $PM_{2.5}$ (percent by mass weight) has been observed across all three episodes (Pre-Diwali, Diwali and Post-Diwali days) except OP, which was found 7-8 times lower association to $PM_{2.5}$ during Diwali-firework days compared to normal and post-Diwali days. During the normal days, Diwali days, and post-Diwali days, chemical determinations accounted for 87.1%, 92%, and 85% respectively, of the gravimetric $PM_{2.5}$ mass. Mg^{2+}, K, Cl^{-}, Se, Ca^{2+}, Sb and Pb have been calculated as source markers of ambient $PM_{2.5}$ measured during crackers burning on Diwali days which are nearly similar to those reported in previous studies. It was gross estimated that firework approximately contributed 32% aerosols load at sampling sites on Diwali day demonstrating the festival as a significant pollution episode.

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APPENDIX

(I) RESEARCH PAPER PUBLISHED

1. **Shippi Dewangan**, Rajan Chakrabarty, Barbara Zielinska and Shamsh Pervez, 2013. Emission of volatile organic compounds from religious and ritual activities in India. ***Environmental Monitoring and Assessment*** 185 (11), 9279-9286.

2. Rajan Chakrabarty, Shamsh Pervez, Judith C. Chow, John G. Watson, **Shippi Dewangan**, Jerome Robles and Guoxun Tian, 2014. Funeral Pyres in South Asia: Brown Carbon Aerosol Emissions and Climate Impacts. ***Environmental Science and Technology Letters*** 1 (1), 44-48.

3. **Shippi Dewangan**, Shamsh Pervez, Rajan Chakrabarty and Barbara Zielinska, 2014. Uncharted sources of particle bound polycyclic aromatic hydrocarbons from South Asia: Religious/ritual burning practices. ***Atmospheric Pollution Research*** 5 (2), 283-291.

4. Shamsh Pervez, **Shippi Dewangan**, Rajan Chakrabarty, Barbara Zielinska, 2014. Indoor VOCs from Religious and Ritual Burning Practices in India. ***Aerosol and Air Quality Research*** 14 (5): 1418–1430.

5. Shamsh Pervez, Rajan Chakrabarty, **Shippi Dewangan**, John G. Watson, Judith C. Chow, Jeevan Lal Matawle and Yasmeen Pervez, 2014. Cultural and Ritual Burning Emission Factors and Activity Levels in India. ***Aerosol and Air Quality Research*** doi: 10.4209/aaqr.2014.01.0022

(II) RESEARCH PAPER COMMUNICATED

1. Shamsh Pervez, Shippi Dewangan, Rajan Chakrabarty, John G. Watson, Judith C. Chow and Jeevan Lal Matawle. Primary assessment of speciated carbonaceous matter and inorganic components of ambient $PM_{2.5}$ during Indian major fireworks festival, Diwali, ***Environmental Geochemistry and Health***.

2. Shippi Dewangan, Rajan Chakrabarty, Shamsh Pervez, John G. Watson, Judith C. Chow, Barbara Zielinska, and Yasmeen Pervez. Study of carbonaceous fractions associated with indoor $PM_{2.5}/PM_{10}$ during Asian cultural and ritual burning practices, ***Aerosol and Air Quality Research.***

(III) RESEARCH PAPER PRESENTED AT NATIONAL/INTERNATIONAL CONFERENCES

1. Shippi Dewangan and Shamsh Pervez, 2014. Indoor Air Quality Status of Respirable Particulate Matter (PM_{10}) in different Indian ritual places. All India Conference (AICON-14), Chhatrapati Shivaji Institute of Technology Durg, 25^{th}-26^{th} April.
2. Shippi Dewangan and Shamsh Pervez, 2014. Study of $PM_{2.5}$ and associated Carbonaceous Fractions in Cultural and Ritual-Indoors. XII Chhattisgarh Young Scientists Congress, Pt. Ravishankar Shukla University Raipur, 17^{th}-19^{th} February.
3. Shippi Dewangan and Shamsh Pervez, 2014. Assessment of Indoor VOCs in different ritual places in Central India. National Conference on Recent Trends in Chemical Sciences, Pt. Ravishankar Shukla University Raipur, 23^{th} -25^{th} January, 2014.
4. Shippi Dewangan and Shamsh Pervez, 2013. Emission estimates of particulate PAHs from religious and ritual burning practices in India. International Congress of Environmental Research (ICER-13), Aurangabad, Maharashtra, 19^{th}-21^{st} December.
5. Shippi Dewangan and Shamsh Pervez, 2013. Emission Factor of Organic and Elemental Carbon Emitted from Religious and Ritual based Burning Activities in India. International Science Congress Association (ISCA-13), Karunya University, Tamilnadu, 8^{th}-9^{th} December.
6. Shippi Dewangan and Shamsh Pervez, 2013. Temporal variation of indoor VOCs in different religious and ritual places in Central India. Indian Council of Chemists Conference (ICC-13), Dharwad, Karnataka, 28^{th}-30^{th} November.
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(IV) INTERNATIONAL/ NATIONAL WORKSHOPS/SEMINARS ATTENDED

1. International Symposium on Environmental-Analytical & Toxicological Issues of Speciation in Atmospheric Organic Pollutant. Pt. Ravishankar Shukla University, Raipur, 6-7 December, 2012.
2. Prospection and Sustainable use of Medicinal Plants in Chhattisgarh, NCNR, Raipur, 10-11 September, 2012.
3. Interactive workshop on Economic Development and Atmospheric Pollution. NEERI, Nagpur, 6-9 February, 2012.
4. International symposium on Novel Approaches to Air Quality Management and Improvement in India. Pt. Ravishankar Shukla University, Raipur, 5-7 September, 2011.
5. 47th Annual Convention of Chemist-2010 & International Conference on Recent Advances in Chemical Sciences. Pt. Ravishankar Shukla University, Raipur, 23-27 December, 2010.

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Emission of volatile organic compounds from religious and ritual activities in India

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Abstract Worshipping activity is a customary practice related with many religions and cultures in various Asian countries, including India. Smoke from incense burning in religious and ritual places produces a large number of health-damaging and carcinogenic air pollutants include volatile organic compounds (VOCs) such as formaldehyde, benzene, 1,3 butadiene, styrene, etc. This study evaluates real-world VOCs emission conditions in contrast to other studies that examined emissions from specific types of incense or biomass material. Sampling was conducted at four different religious places in Raipur City, District Raipur, Chhattisgarh, India: (1) Hindu temples, (2) Muslim graveyards (holy shrines), (3) Buddhist temples, and (4) marriage ceremony. Concentrations of selected VOCs, respirable particulate matter (aerodynamic diameter, <5 μm), carbon dioxide, and carbon monoxide were sampled from the smoke plumes. Benzene has shown highest emission factor (EF) among selected volatile organic compounds in all places. All the selected religious and ritual venues have shown different pattern of VOC EFs compared to laboratory-based controlled chamber studies.

Keywords Emission factor · Volatile organic compounds · Indoor air pollution · Religious and ritual places

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Introduction

Volatile organic compounds (VOCs) participate in the atmospheric photochemical reactions forming secondary organic aerosol on oxidation (Platnick and Twomey 1994; Feingold and Morley 2003) and ozone in the presence of NOx (Pun et al. 2002; Pinto et al. 2007). It has been estimated that environmental damage due to the precursors of O_3 (VOC and NOx) emissions is roughly $1,000/Mg VOCs (Rable and Eyre 1998). The adverse health effects of certain VOC include chronic obstructive pulmonary disease, bronchial asthma, systolic plus diastolic hypertension, and lung cancer (Guo et al. 2004; Arif and Shah 2007). Selected VOCs have been quantified in various environments including ambient air (Kumar and Viden 2007; Srivastava et al. 2006); residential–indoor air (Beck et al. 2007), workplace–indoors (Tovalin-Ahumada and Whitehead 2007), and personal exposures (Tovalin-Ahumada and Whitehead 2007; Saborit et al. 2009). Apart from atmospheric VOC studies, emission estimation from different

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Funeral Pyres in South Asia: Brown Carbon Aerosol Emissions and Climate Impacts

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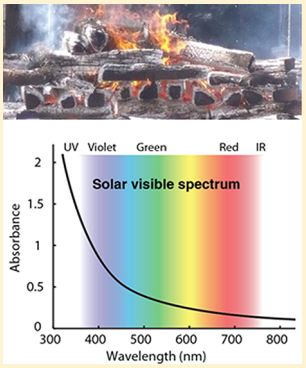
Supporting Information

ABSTRACT: Atmospheric heating caused by anthropogenically emitted carbonaceous aerosols contributes to one of the largest uncertainties in climate forcing over south Asia (SA). Past studies have identified the combustion of fossil fuels and residential biofuels as being the dominant emitter of light-absorbing black carbon aerosols over this region. Here, we measure emissions from open-air burning of funeral pyres, a deep-rooted and widely prevalent custom in SA, and find that large amounts (≈98% by mass) of light-absorbing organic carbon (OC) aerosols, optically defined as brown carbon (BrC), are emitted per kilogram of feedstock burned. The emitted OC contributes an average 40% to the smoke particulate matter absorption of the visible solar radiation. We calculate funeral pyres in SA contribute approximately 92 Gg of light-absorbing OC annually, which is equivalent to ≈10 and 23% of the carbonaceous aerosol mass from regional biofuels and fossil fuels, respectively. Our findings underscore the importance of accounting for cultural burning practices as aerosol sources in emission inventories and BrC aerosols in climate models, as well as the development of mitigation strategies.



INTRODUCTION

South Asia (SA) is one of five regional hot spots for emitting anthropogenic carbonaceous aerosol into the atmosphere.[1] Black carbon (BC) and organic carbon (OC) aerosols in the atmosphere over this region alter net solar radiative flux to the Earth's surface, the net effect of which may outweigh the regional warming effect of greenhouse gases.[2] BC aerosols are emitted as byproducts of incomplete combustion of fossil fuels and biofuels. BC aerosols absorb sunlight in visible wavelengths, increasing inversely with wavelengths from the near infrared (1 μm) to the ultraviolet with a power law of 1.[3] Their light absorbing characteristics have been implicated in regional atmospheric warming,[2] changing Asian monsoon patterns,[4] and accelerated melting of the Himalayan glaciers.[2] OC aerosols are emitted both from high- and low-temperature combustion systems in primary and secondary forms; the latter are produced by atmospheric oxidation of co-emitted volatile organic compound gases.[3,5] OC aerosols absorb sunlight insignificantly at visible wavelengths,[6,7] with the exception of brown carbon (BrC), a class of OC, which strongly absorbs in the short visible wavelengths, relative to BC.[3] BC and OC are always present in combustion-generated particles in varying proportions.

The aerosol radiative effects over the SA region represent one of the largest uncertainties in climate models.[1] This uncertainty results from our inadequate knowledge of regional emission source types, emission rates, and associated aerosol microphysical properties.[8] Recent studies have suggested that current emission inventories (EIs) may be underestimating emissions of light-absorbing carbonaceous aerosols by a factor of 2−3.[9] During the past decade, compilers of aerosol EIs over SA primarily have focused on combustion sources used to meet energy requirements of the region's rapid economic growth and technology development.[8,10,11] Consequently, burning of fossil fuel and residential biofuels have been recognized as the two primary energy-producing combustion sources.[12,13] However, a large discrepancy in source apportionment of BC and OC in this region still exists. Ambient measurements in SA suggest that between 30 and 90% of light-absorbing carbonaceous aerosols originate from fossil fuel burning, while EI compilers, relying on annual fuel consumption and laboratory-based emission factors (EFs), suggest that between 20 and 50% of emissions can be attributed to fossil fuels.[14] Aerosol EFs, defined as grams of particulate mass emitted per kilogram of dry fuel burned, obtained from laboratory-based measurements differ significantly from those obtained in real-world emission scenarios.[8,11,15] Significant differences also have been observed between laboratory and real-world measurements of aerosol light absorption properties.[8,16] Here, we investigated aerosol emissions from a hitherto unstudied but widely prevalent pollution source in SA, open-air funeral pyre cremation (FPC), and found that these emissions have a measurable effect on the aerosol mass budget and radiation balance.

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Uncharted sources of particle bound polycyclic aromatic hydrocarbons from South Asia: Religious/ritual burning practices

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ABSTRACT

This paper deals with the real–world emissions of particle bound polycyclic aromatic hydrocarbons (PAHs) emitted from different religious and ritual burning practices in India. The emission factors (EFs) of various PAHs were characterized over integrated stages of burning practices including pyrolysis, flaming and smoldering. Sampling was carried out in selected five different types of religious and ritual places: Hindu Temples (HT), Muslim Holy shrines (MG), Buddhist Temples (BT), Hindu Marriage Places (MP) and Cremation Centers (Hindu Funeral Pyre) (CC); where burning practices mostly include natural and synthetic biomaterials in different proportions. Seventy PAHs were analyzed and grouped in light PAHs (LPAHs=25) and heavy PAHs (HPAHs=45) according to number of benzene rings present and respective molecular weights. The average EF of total PAHs (TPAHs) from MP, MG, BT, HT and CC is estimated as 77.04±5.39, 99.09±6.02, 90.35±6.76, 22.78±1.57, and 77.15±6.16 mg kg^{-1}, respectively. Country level emission budget of particulate bound TPAHs from religious and ritual burning practices has also been evaluated to 0.47 Gg yr^{-1}; contributed ~23% of the 2.07 Gg yr^{-1} – Indian emission budget of PAHs from unknown sources reported earlier.

Keywords: *Polycyclic aromatic hydrocarbons, religious and ritual burning practices, emission factor, emission budget*

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1. Introduction

The sources, formation, fate and pathways of atmospheric polycyclic aromatic hydrocarbons (PAHs) released from different burning practices of biomass have received major attention over the world (Chang et al., 2009; Yang et al., 2012; Mu et al., 2013; Singh et al., 2013). Most PAHs are produced by incomplete combustion and pyrolysis of organic substances during different burning processes. PAHs are of special interest due to their toxicity, carcinogenicity, and ubiquitous distribution in the environment (Khalili et al., 1995; IARC, 2002). Burning incense sticks in the home and temples is an important religious and ritual activity in Oriental society and emit significant quantities of PAHs (Schoenotol and Gibbard, 1967; Chiang and Liao, 2006; Yang et al., 2012). Many individual PAHs are cytotoxic and mutagenic to organisms especially benzo[*a*]anthracene, benzo[*k*]fluoranthene and benzo[*a*]pyrene, indeno[1,2,3–*cd*]pyrene have been recognized as potential carcinogens for humans (ATSDR, 1995). Several studies have reported high exposure levels of the organic compounds such as PAHs and total suspended particle (TSP), particularly, in poorly ventilated houses and exposed subjects have shown higher rate of cellular DNA destruction, and impaired regenerative capability (Naumova et al., 2002; Oanh et al., 2002; Lung and Hu, 2003; Ohura et al., 2004; Yang et al., 2006; Yang et al., 2007; Navasumrit et al., 2008).

Several studies have been conducted worldwide to determine emission factors (EFs) of PAHs from combustion activities involved with domestic and industrial processes using rice straw, agricultural crop residue, fossil fuels (coal, fuel woods and dung cakes) (Oanh et al., 1999; ICMR, 2001; Venkataraman et al., 2002; Singh et al., 2013), transportation, forest fires and cooking (Dubowsky et al., 1999; Fischer et al., 2000; Gadi et al., 2012), burning of natural gas, liquefied petroleum gas and kerosene (Rogge et al., 1993; Mitra and Ray, 1995; ICMR, 2001) and candles and incense burning (Lau et al., 1997; Li and Ro, 2000). The study described here is focused on development of real–world EFs of selected PAHs from burning practices involved with major religious and ritual activities; mostly occur in South Asian countries. This study is the extended part of the comprehensive assessment of emission characterization of organic aerosols and volatile organic compounds from religious and ritual burning practices in India (Chakrabarty et al., 2013; Dewangan et al., 2013). Annual emission estimates of PAH emissions from religious and ritual burning practices in India was also described.

2. Materials and Method

2.1. Sampling plan

To determine the EFs of selected PAHs, a grab sampling plan in selected religious/ritual burning practices occurred in Raipur city located in Chhattisgarh, India has been adopted (Lin et al., 2002; Masih et al., 2010; Chakrabarty et al., 2013; Dewangan et al., 2013). Raipur, the capital city of Chhattisgarh, India, is located in global scale of 21°14′22.7″N latitude and 81°38.1″E longitude (Figure 1). According to Census India (2011) population of Raipur Municipal Corporation was about 10 million. Raipur has a tropical

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Indoor VOCs from Religious and Ritual Burning Practices in India

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ABSTRACT

Measurements of selected volatile organic compounds (VOCs), known as markers of emissions resulting from burning practices using natural- and synthetic biomaterials, have been carried out in major religious/ritual-indoors in India. Four different religious/ritual-indoors were selected for monitoring purposes: 1) Hindu Marriage Places (MP), 2) Muslim Holy Shrines (MG), 3) Buddhist Temples (BT), and 4) Hindu Temples (HT). One pure residential-indoor (RESID) site was also examined for comparison studies. Indoor VOCs sampling was carried out throughout the three seasons of summer, fall and winter of the calendar year of 2012–2013 in Raipur, India. VOCs samples, collected by passive sampling over a 48-h period, were analyzed using thermal desorption (TD), followed by high-resolution gas chromatographic separation and mass spectrometric detection (GC/MS). A total of 14 volatile organic compounds (n-hexane, cyclohexane, n-heptane, n-octane, n-nonane, n-decane, n-undecane, styrene, o-xylene, m,p-xylene, 1,2,4-trimethyl benzene, ethylbenzene, benzene, and toluene.) were quantified. The annual mean concentrations for total VOCs (TVOCs) were 216.61 ± 75.15, 656.34 ± 220.82, 681.75 ± 219.83, 129.51 ± 45.24 and 82.67 ± 40.96 μg/m^3 for MP, MG, BT, HT and RESID respectively. The results were found to be higher than the prescribed standards and earlier reported indoor VOCs levels. Indoor/outdoor ratios (I/O), correlation analyses, seasonal variations and indoor/outdoor contributions to the measured levels are also investigated.

Keywords: VOCs; Indoor air quality; Religious and ritual places; Indoor/Outdoor ratio; Seasonal variation.

INTRODUCTION

Volatile organic compounds (VOCs) have now been identified as one of the major indoor air contaminants due to construction of well-sealed houses using various types of insulators (Berk *et al.*, 1980), usage of painting materials in indoor-houses (Srivastava *et al.*, 2000), household cooking fuels (Liquid petroleum gas, kerosene, solid fuels etc.) (Sinha *et al.*, 2006; Huang *et al.*, 2011), adhesive- bonded material use in plywood/ PVC flooring (Low *et al.*, 1998; Wilke *et al.*, 2004; Jarnstom *et al.*, 2008; Choi *et al.*, 2010; Kang *et al.*, 2013) and house furniture's (Fisher *et al.*, 1962; NRC, 1980; USDHUP, 1980; Franklin, 1981). Furthermore, Selected VOCs emission over the building materials has also been reported by Molhave (Molhave, 1980). High lipid solubility character of these VOCs explained that they are rapidly absorbed through the lungs and enter organs having high content of lipids. VOCs are also reported that they rapidly cross the blood-brain barrier and commonly depress central nervous system and cardiac functions (Beall and Ulsamer, 1980). Human exposure to one of the VOCs namely n-hexane causes demyelination and degeneration of peripheral nerves (Paulson and Wylonis, 1976; Spencer *et al.*, 1980). Similarly toluene which was observed to be present in more than 50% of indoor air samples, causes fatigue, muscle weakness, and confusion in humans exposed for eight hours to atmospheric concentrations of 200 to 300 ppm (Berk *et al.*, 1980; Benett and Forman, 1980; Konietzko *et al.*, 1980). Benzene and toluene vapors have been reported to cause central nervous system depression, psychosis, and liver and kidney damage (Aruffo and Escobar, 1979; Tarsh, 1979); benzene also damages the hematopoietic system, causing first a decrease in the number of white blood which manifests themselves as anemia, leukopenia, thrombocytopenia, and leukemia (Beall and Ulsamer, 1980).

Most of air quality studies on VOCs status conducted in Indian sub-continent are mainly focused on ambient-outdoor environment (Rao *et al.*, 1996; Chattopadhyay *et al.*, 1997; Srivastava, 2004; Srivastava *et al.*, 2004, 2006; Dutta *et al.*, 2009) and fewer indoor air quality studies were reported, specifically for residential cooking activity emissions (Raiyani *et al.*, 1993; Mishra, 2003; Pandey *et al.*, 2005;

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Cultural and Ritual Burning Emission Factors and Activity Levels in India

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ABSTRACT

Real-world particulate matter, organic carbon, and elemental carbon (OC and EC) emission measurements were measured for different cultural and ritual services. These were (g/kg): 11.36 (OC), 0.27 (EC) and 31.04 (RPM) of Marriage Events; 27.04 (OC), 0.18 (EC) and 123.82 (RPM) of Muslim Holy Shrines; 25.99 (OC), 0.85 (EC) and 47.93 (RPM) of Buddhist Temples; and 3.47 (OC), 7.96 (EC) and 20.13 (RPM) of Hindu Temples. When projected to reasonable levels of these cultural/ritual activity levels in India, total annual emissions would be 72.38 Gg/yr, comparable to those from transport (165 Gg/yr), power plant (19 Gg/yr), agricultural waste burning (428 Gg/yr) and forest and savanna burning (176 Gg/yr).

Keywords: Emission factors; OC and EC fractions; Cultural and ritual performances; Combustion activities; Asian Brown Haze.

INTRODUCTION

"Brown Clouds" are caused by a mixture primary and secondary particulate matter (PM) generated from engine exhaust, biomass burning, and industrial processes (Smith *et al.*, 1983; Reiner *et al.*, 2001; Streets *et al.*, 2003; Yan *et al.*, 2006; Witham and Manning, 2007). Atmospheric elemental carbon (EC) and organic carbon (OC) are the major primary PM constituents emitted by inefficient combustion of fossil fuels and biomass (USEPA, 2012; UNEP and NOAA, 2003). EC and OC occur together in PM as soot, and their combination is important for climate change because EC absorbs solar radiation, potentially contributing to global warming (Jacobson, 2001; Jacobson, 2002; Hansen and Nazarenko, 2004; Bond and Sun, 2005; Ramanathan *et al.*, 2007). However, EC/OC soot also scatters solar radiation back into space, which may counteract the warming effect. Water-soluble OC may be mixed in liquid water, thereby modifying its optical properties and affecting the formation and surface tension of cloud droplets (Hallett *et al.*, 1989; Saxena *et al.*, 1995; Rivera-Carpio *et al.*, 1996; Saxena and Hildemann, 1997; Soto-Garcia *et al.*, 2011).

In India, large amounts of EC and OC are emitted from agricultural crop residue burning (Streets *et al.*, 2003; Vadrevu *et al.*, 2008; Gadde *et al.*, 2009; Rehman *et al.*, 2011; Sahai *et al.*, 2011), fossil fuel combustion in residential (Raiyani *et al.*, 1993; Kandpal *et al.*, 1995), industrial sectors (Sharma and Vyas, 2001; Ghose and Majee, 2003; Mishra, 2004), and transportation (Ghose *et al.*, 2004; Pal *et al.*, 2009; Nesamani, 2010; Apte *et al.*, 2011; Reynolds *et al.*, 2011), forest fires (Vadrevu *et al.*, 2006; Badarinath *et al.*, 2007; Vadrevu *et al.*, 2011) and other activities. EC and OC emission factors from religious and ritual burning activities have not been adequately characterized, nor have the amounts of material burned each year been estimated. This study measures these emissions factors, quantifies the activity levels, and estimates contributions to brown cloud formation for several types of Indian religious rituals.

There are ~3.0 million religious worship places in India, more than the number of educational and health centres in India (Ministry of Health and Family Welfare, 2011; Office of registrar general, 2012). In a Hindu marriage, the couple transits seven circuits around a Holy Fire fuelled by the materials described elsewhere (Dewangan *et al.*, 2013, 2014). Besides these ritual activities, different religion oriented worship places (Hindu Temples, Muslim Holy Shrines and Buddhist Temples) are also paid spiritual homage using flaming episodes by igniting different kinds of bio- and synthetic materials described elsewhere (Dewangan *et al.*, 2013, 2014). This study evaluates real-world emission conditions in contrast to other studies that examined emissions from specific types of incense (Lee and Wang, 2004; See and Balasubramanian, 2011) or biomass material (Yevich

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